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Fuel-Neutral Studies of Particulate Matter Transport Emissions

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Pacific Northwest National Laboratory

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Overview

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Timeline

- Start - FY16*
- Finish - FY18

* Three-year scope proposed in response to 2015 National Laboratory FOA

Budget

- Funding received in FY16 - $250K
- Planned budget for FY17 - $250K

Barriers

- Barriers addressed for enabling of high-efficiency engine technology:
  - B.** Lack of cost-effective emission control
  - C.** Lack of modeling capability for combustion and emission control
  - F.** Lack of actual emissions data on pre-commercial and future combustion engines

** Indexed to list in VTO Multi-Year Program Plan

Partners

- General Motors Company - provide project guidance, support for ERC
- Engine Research Center at University of Wisconsin, Madison - host and operate test engines, perform experiments
- Massachusetts Institute of Technology - Micro X-Ray CT

Relevance and objectives

Overall objective: Enable adoption of future high-efficiency engine technologies

2016 Chevy Cruze with 1.4 L turbocharged DI LE2 engine
By Ryan Hildebrand - Own work, CC BY-SA 4.0,
https://commons.wikimedia.org/w/index.php?curid=46673960

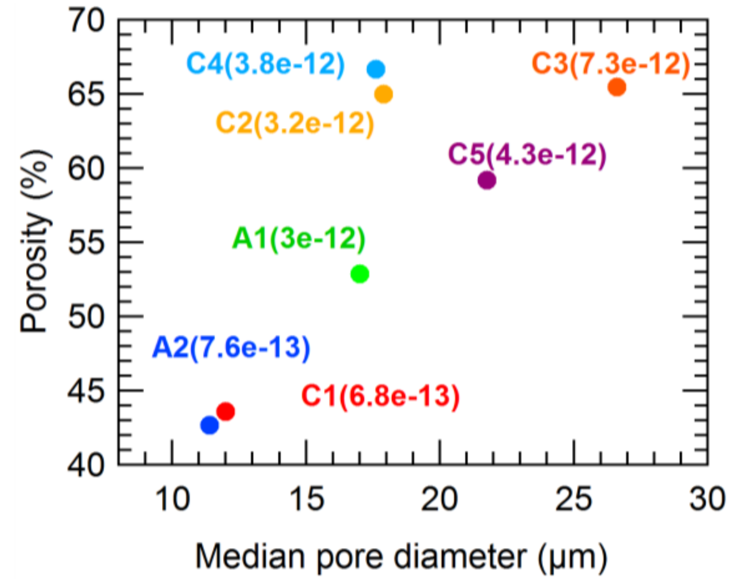
Barrier: Lack of actual emissions data on pre-commercial and future combustion engines
Objective: Comprehensive particulate characterization with single-cylinder test engines, guided by industry

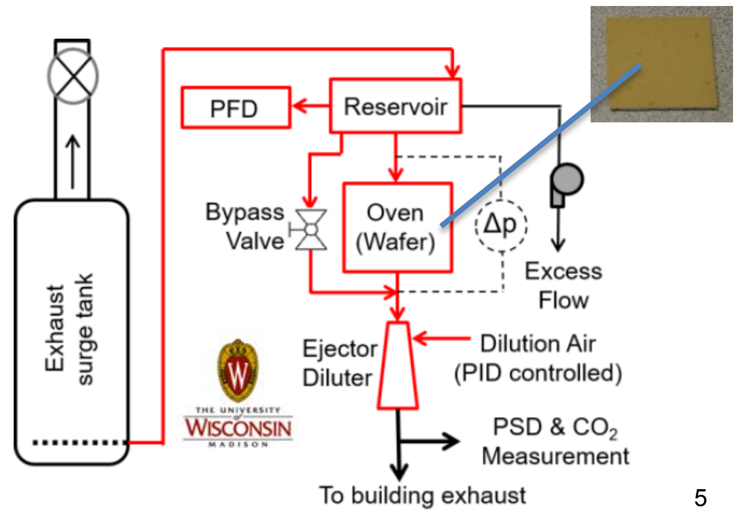
Barrier: Lack of cost-effective emission control
Objective: Seek to shorten development time of filtration technologies for future engines by improving fundamental understanding of how filter media properties impact back-pressure and filtration efficiency

Barrier: Lack of modeling capability for combustion and emission control
Objective: Develop modeling approaches relevant to the likely key challenge for SIDI filtration – high number efficiency at high exhaust temperatures (implying little soot accumulation in filters)

Fundamental experiments

- Detailed SIDI particulate characterization (previous stages of this collaboration)
  - Characterization of exhaust particulates over a wide range of fuels and operating conditions
  - Fundamental studies around soot formation
- Filter characterization (see backup slide for more detail)
  - Mercury porosimetry
  - Capillary flow porometry
  - Micro X-Ray CT
  - Pore network modeling
  - Micro-scale lattice-Boltzmann simulations
- Exhaust Filtration Analysis (EFA) filtration experiments at U of Wisconsin, Madison
  - Wide variety of filters and particulate populations
  - Focus on low (but non-zero) soot loadings
  - Low temperature experiments
  - High temperature experiments (continuous regeneration)





Moving toward improved filter models

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- Spherical unit collector models represent the solid structure of filters as spheres with shells that fill uniformly with soot
  - Parameters typically tuned to match experimental data
  - Successful in design of DPF systems
  - Difficult to predict performance over a wide range of filter properties - especially for the very small particles that dominate in gasoline exhaust
- Extensions have been proposed in previous presentations
  - Alternate diffusion term
  - Distribution of collector sizes (U of Wisconsin HMF model - Gong and Rutland)
- More realistic alternatives such as the constricted tube model may be more widely applicable
  - Inform using new methods for characterizing filter microstructure
  - Validate with extensive set of EFA filtration data

> *"All models are wrong. Some models are useful."*
> *George E. P. Box*

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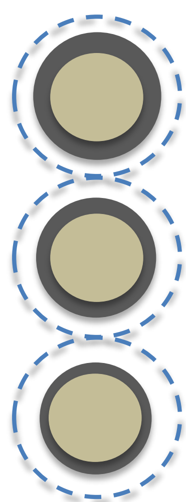
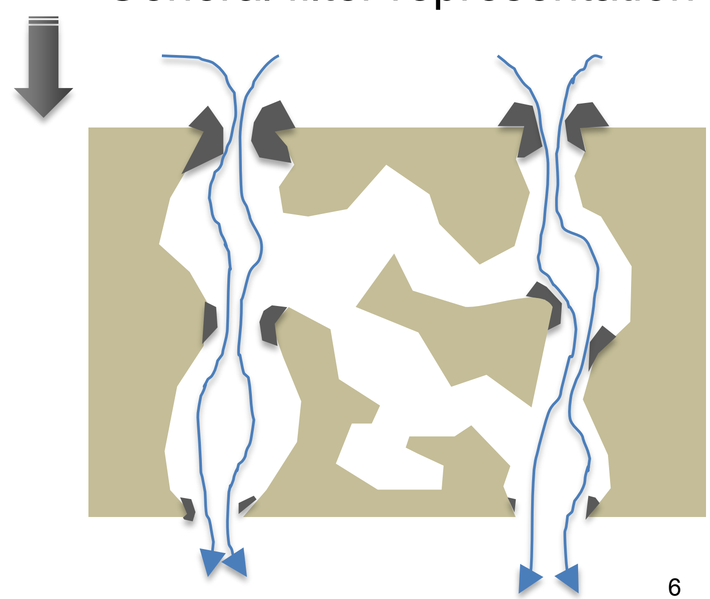
Spherical unit collector model



General filter representation



Gong, J. and C. J. Rutland (2015). "PDF-Based Heterogeneous Multiscale Filtration Model." Environmental Science & Technology **49**(8): 4963-4970.

Technical accomplishments
Filter characterization

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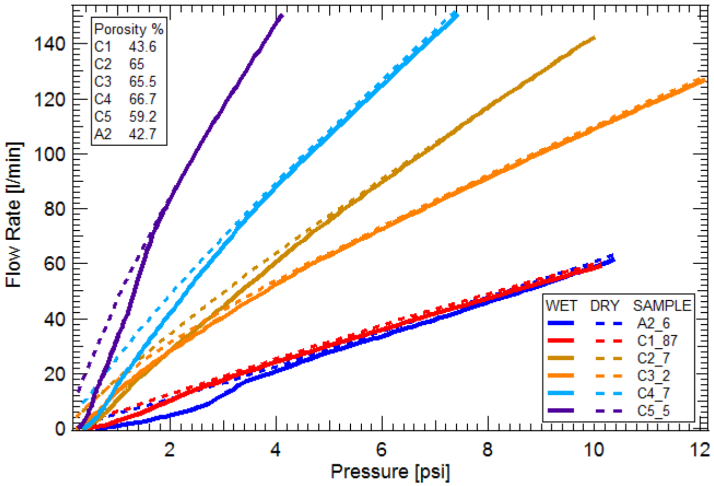
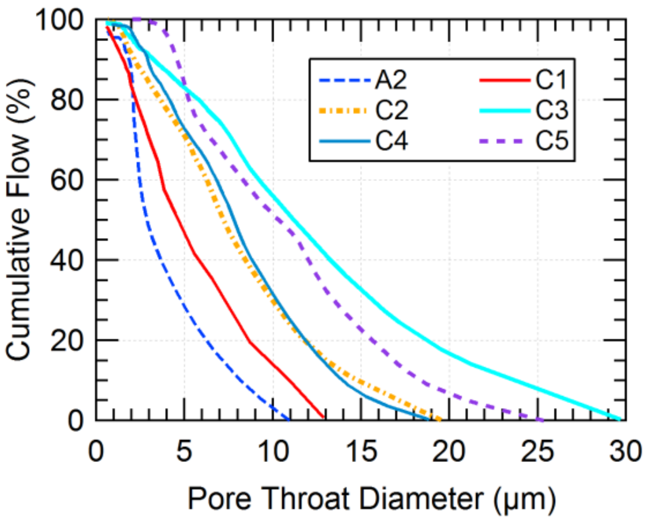
Capillary Flow Porometry

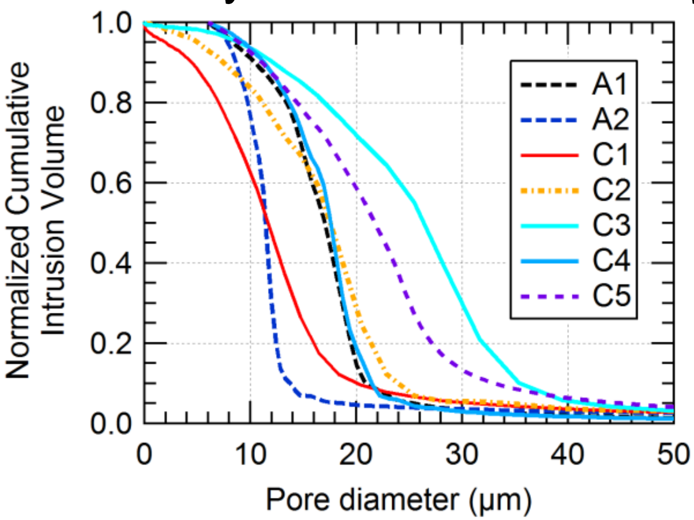


- Capillary flow porometry (CFP) is a technique which uses a gas at progressively higher pressures to displace a wetting liquid from the porous filter samples
- Unlike mercury intrusion porosimetry (MIP), CFP targets pores extending through the filter wall
- Minimum throat diameters along flow paths are identified, as well as flow carried by each size of pore
- A third method, liquid extrusion porosimetry, was also evaluated, but did not cover the relevant pore size range

CFP provides complementary information to MIP



Mercury Intrusion Porosimetry

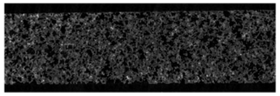


X-Ray CT artifacts, segmentation methods

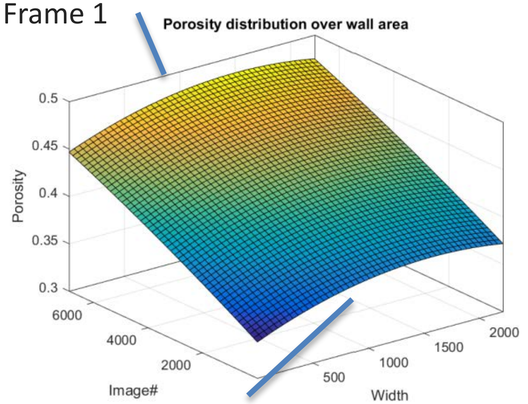
- Segmentation is the process of reconstructing solid models from X-Ray CT data
- Grayscale thresholding is the simplest method
- Artifacts such as beam hardening led to unrealistic distributions of porosity over filter wall areas
- Many artifacts can be reduced or compensated for by the X-ray operator - it pays to be an informed customer
- More advanced segmentation methods including the “top hat” and “water shedding” approaches were explored

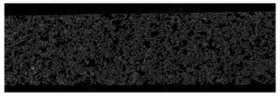
Current solution: custom grayscale adjustment was used to obtain better reconstructions from the existing data

Data for C1 had a darkness gradient from one end of the specimen to the other



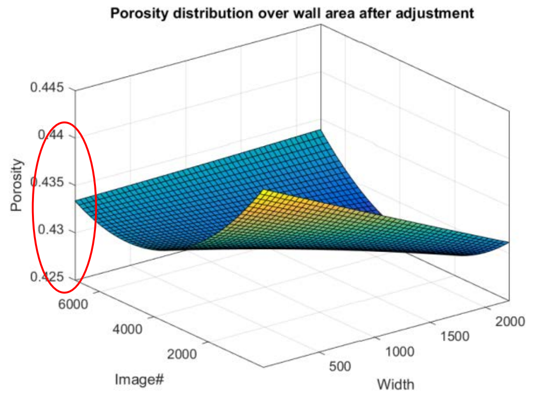
Frame 1



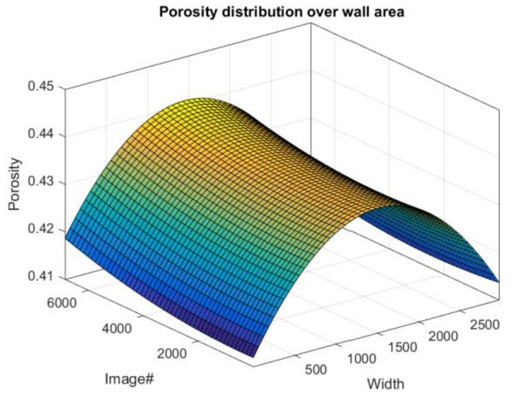


Frame 6904

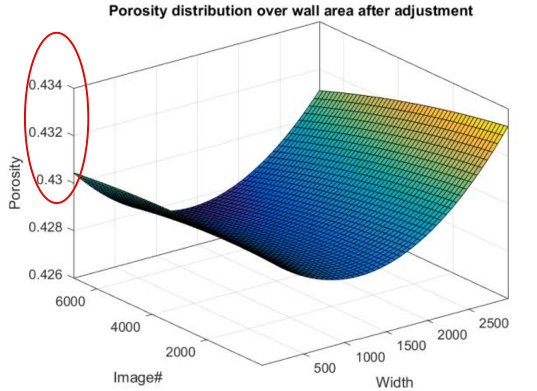
Adjustment



Data for A2 had typical beam hardening artifacts (darker in center)



Adjustment



All dimensions in voxels = 1.67 μm

Pore network modeling

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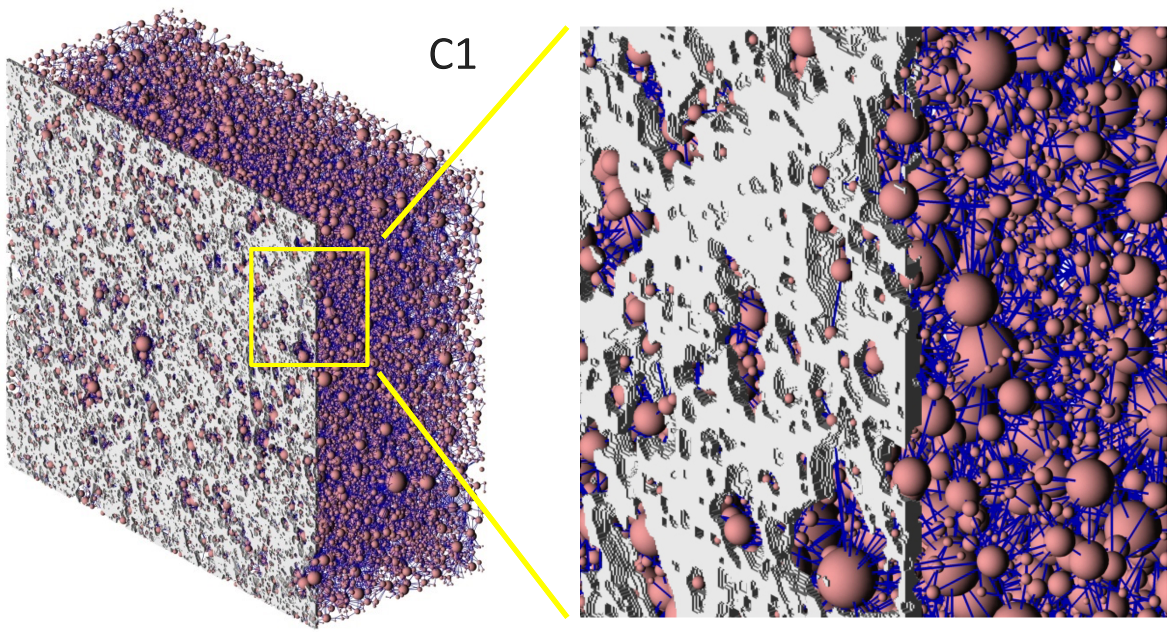
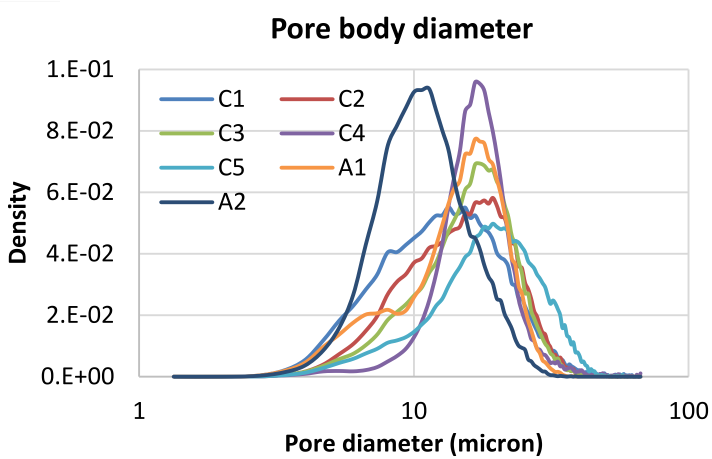
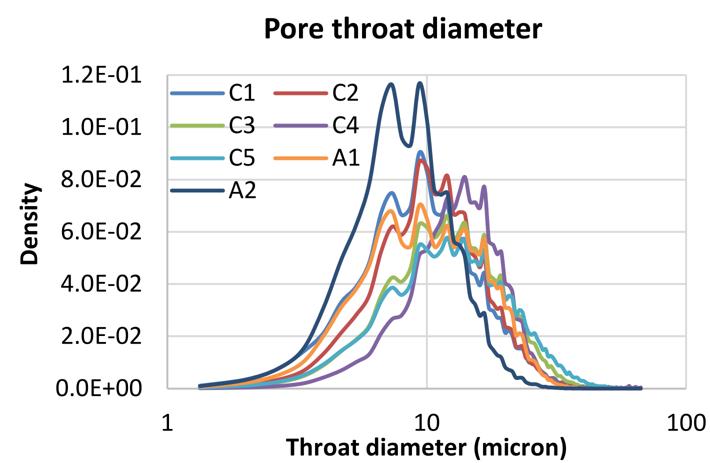


Comparison between original geometry and pore network (only a thin slice of original geometry is shown)

- Pore network models were created for each substrate using the maximal inscribed sphere method
- Provide details about pore connectivity, pore throat vs. body dimensions
- The resulting pore networks can also be use to do approximate permeability estimates

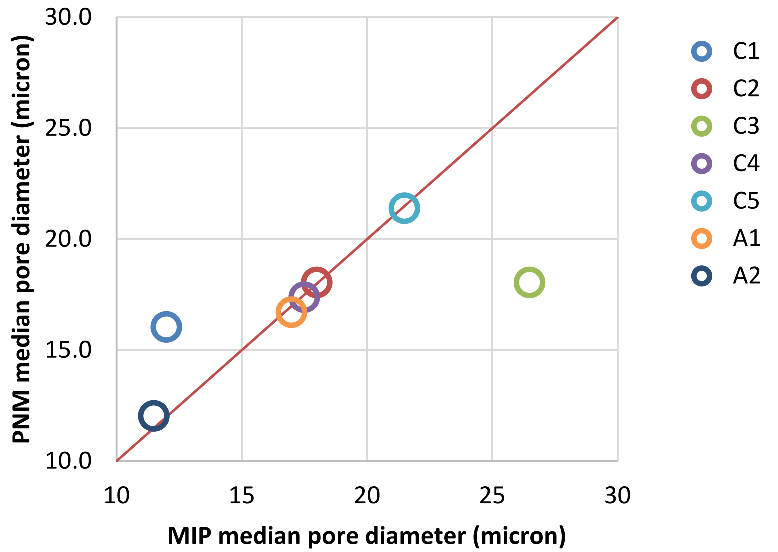




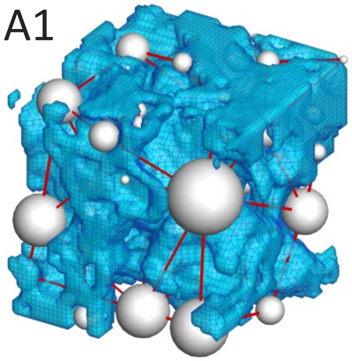
A.S. Al-Kharusi, M.J. Blunt, Journal of Petroleum Science and Engineering 56 (2007) 219-231
H. Dong, M.J. Blunt, Physical Review E 80 (2009) 036307

Geometric parameters from pore networks

- "Pore bodies" are the wide points in the pore networks
- "Pore throats" are the constrictions where soot, ash will accumulate
- Median body diameter seems to correlate most closely with median diameter from Hg porosimetry
- Median pore throat diameters were 67% to 85% of median body diameters
- Throat length is measured between centers of connected pore bodies - may provide useful information for spacing of collectors in wall models



A1



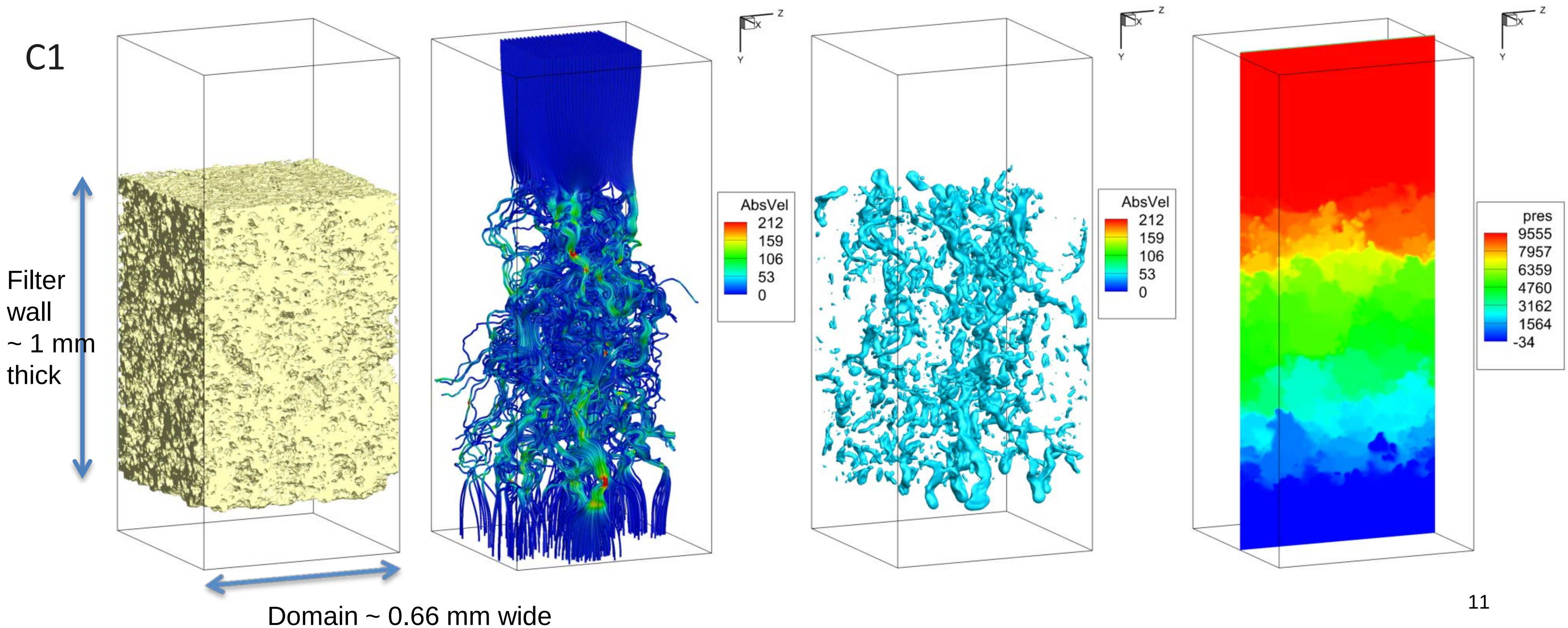
	Hg porosimetry	Pore network model				
	Median diameter (micron)	Median body diameter (micron)	Median throat diameter (micron)	Median throat length (micron)	Throat/ body diameter ratio	Throat length/ Body diameter
C1	12	16.0	11.4	75	0.71	4.7
C2	17.9	18.0	12.0	90	0.67	5.0
C3	26.6	18.0	14.0	100	0.78	5.6
C4	17.6	17.4	14.7	90	0.85	5.2
C5	21.7	21.4	15.4	105	0.72	4.9
A1	17	16.7	12.7	80	0.76	4.8
A2	11.4	12.0	9.4	60	0.78	5.0

Pore network models can provide a wide array of information on pore size, shape, and connectivity

New lattice-Boltzmann simulations

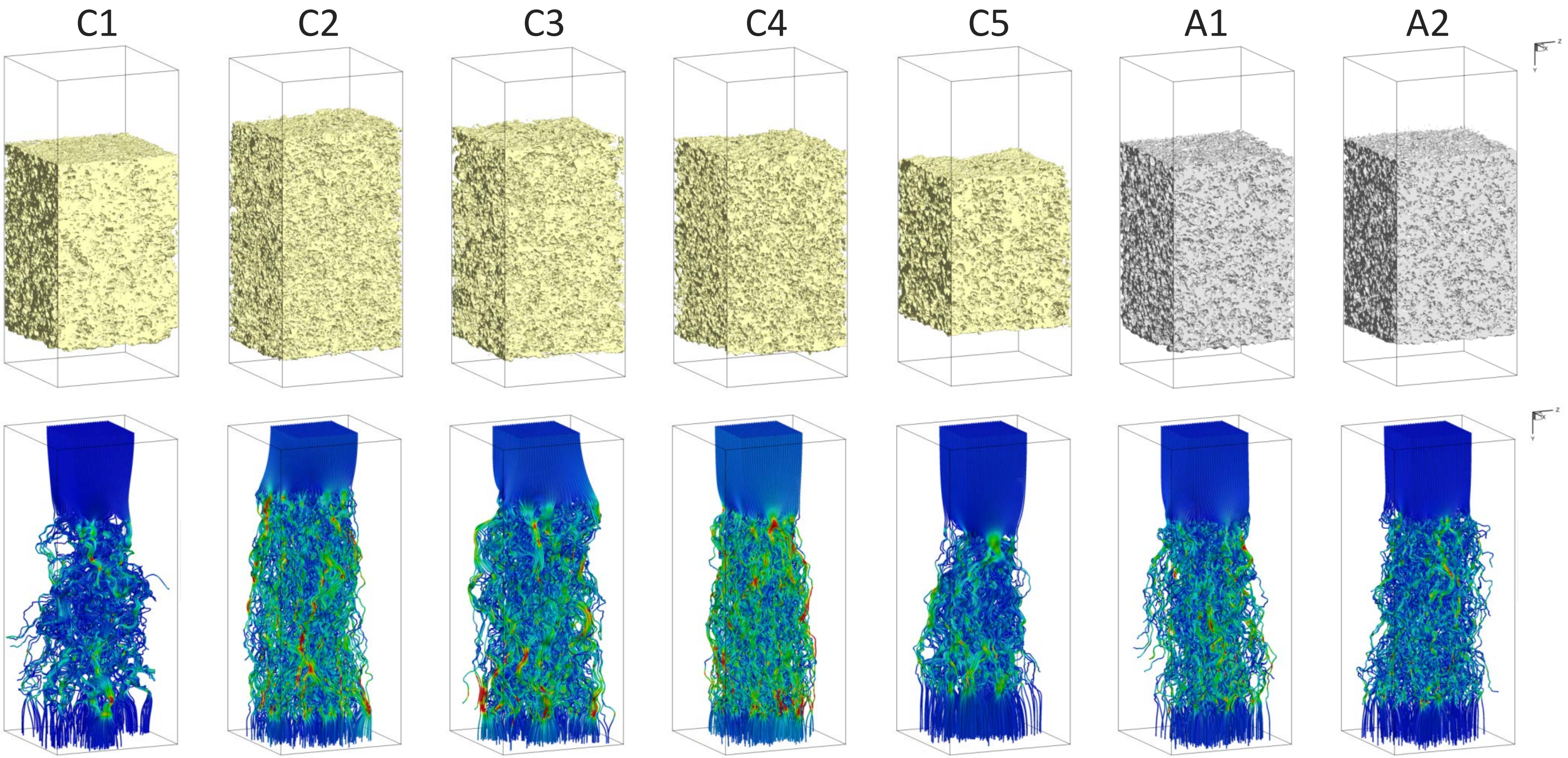
- New series of LB simulations
- Coarsened once from native CT resolution: dx = 3.34 μm
- Somewhat larger computational domains than those shown last year: 200 × dx per side in X and Z = 648 μm
- 19E+06 computational cells
- Periodic boundaries on sides

Gas flow direction



New lattice-Boltzmann simulations

- LB simulations carried out for all seven bare substrates
- Detailed three-dimensional flow field solutions used for subsequent analyses



Technical accomplishments
Tortuosity estimates from 3D reconstructions

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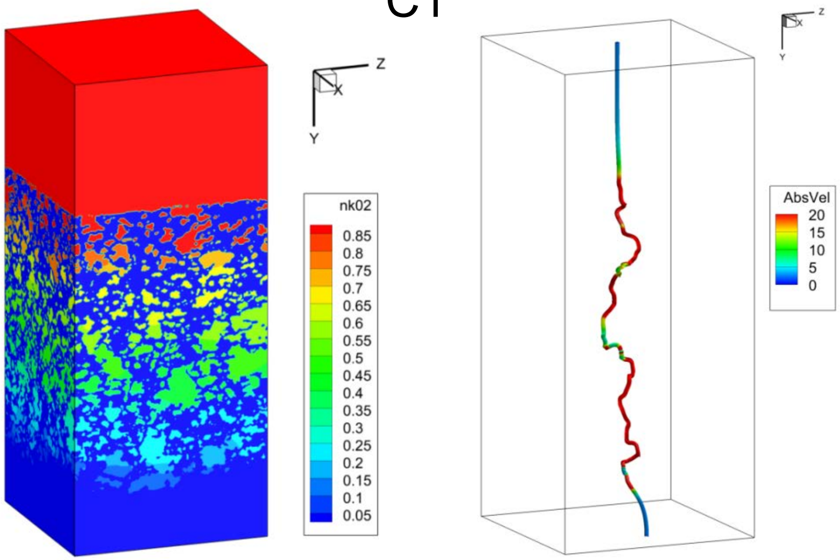
C1



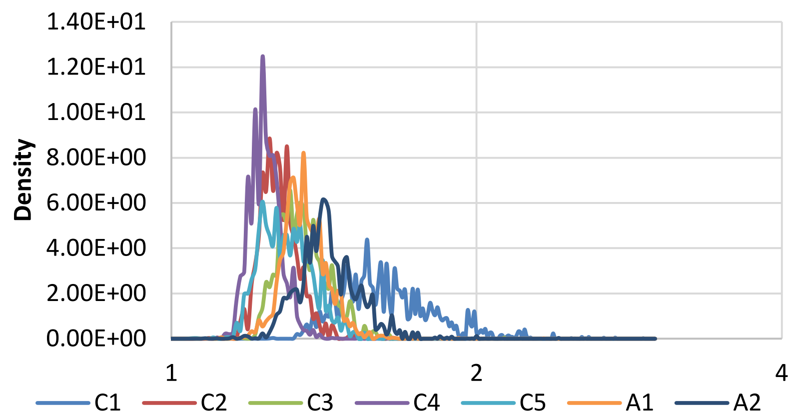
	Porosity	W	Diffusion Tortuosity	Median streamline tortuosity
C1	0.43	0.71	5.9	1.62
C2	0.65	0.3	1.9	1.27
C3	0.65	0.32	2.0	1.34
C4	0.67	0.22	1.8	1.24
C5	0.59	0.52	1.9	1.29
A1	0.53	0.24	2.3	1.35
A2	0.42	0.21	4.1	1.42

- Tortuosity is an important concept in transport through porous media
- Two methods used:
  - Effective diffusion length
  - Average streamline length

$$\tau_{Diff} = \varepsilon \frac{D_{AB}}{D_{eff}}$$

τ_{Diff} - tortuosity factor
ε - porosity
D_{AB} - binary diffusivity
D_{eff} - effective diffusivity

Streamline tortuosity distribution



$$W = \frac{d_{50} - d_{10}}{d_{50}}$$

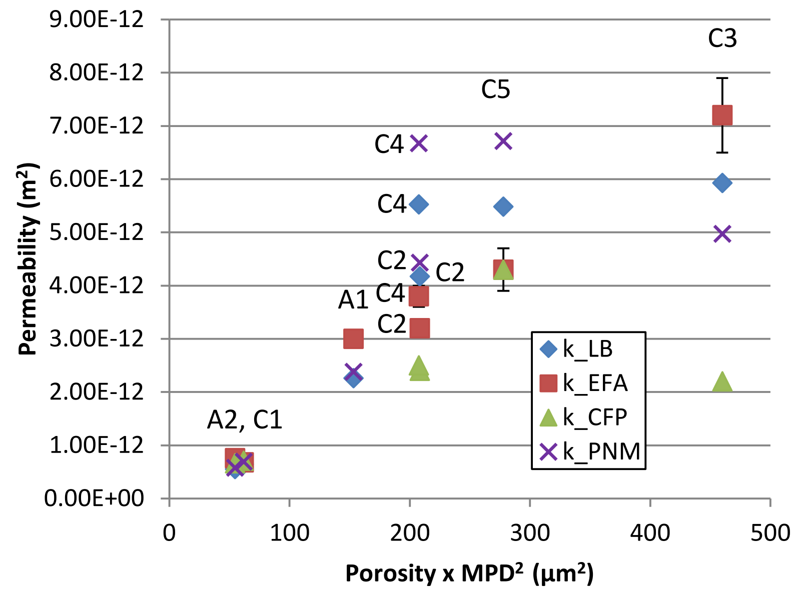
“W” metric provides a measure of mercury pore size distribution width - thought to be related to pore connectivity

Realistic tortuosity is one example of a possible path toward improved device scale models

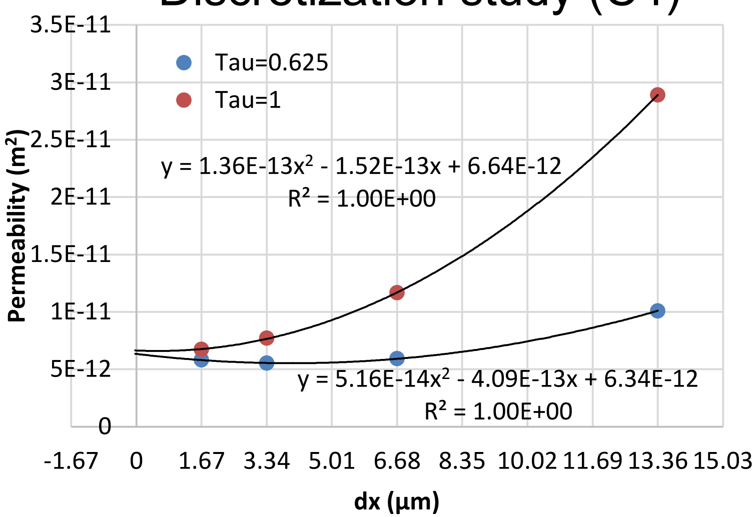
Technical accomplishments

Permeability predictions, LB checks

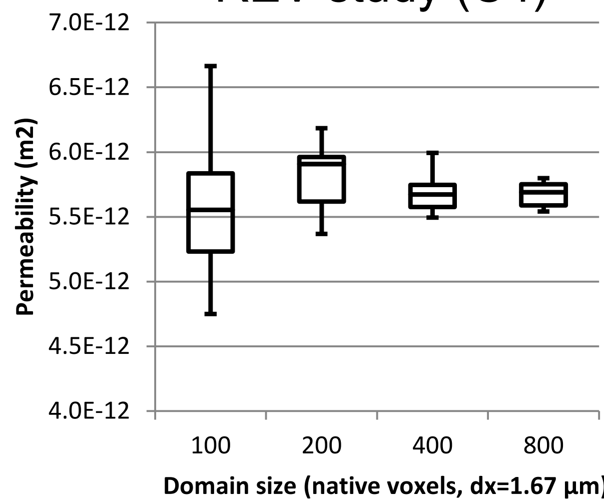
- Permeability predictions made using various methods were not identical, but showed similar trends
  - CFP and PNM suggested smaller pores for C3 than measured by Hg porosimetry → lower permeability
  - All methods agreed that C4 is more permeable than C2
- Discretization study suggests that 3.3 μm resolution is adequate for bare substrates with LB relaxation parameter (tau) = 0.625
- Representative equivalent volume (REV) analysis: larger domains for LB simulations probably would not dramatically change predictions
- Methods other than EFA are impacted more by sample-to-sample variation: smaller volumes, fewer specimens



Discretization study (C4)

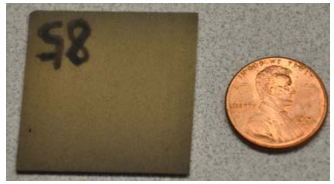


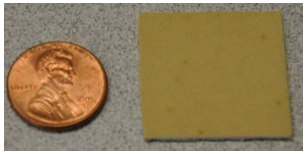
REV study (C4)

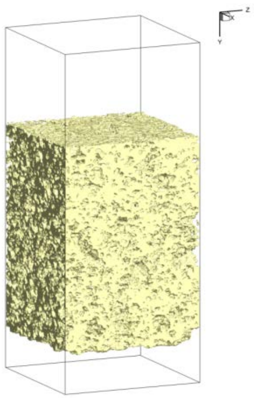


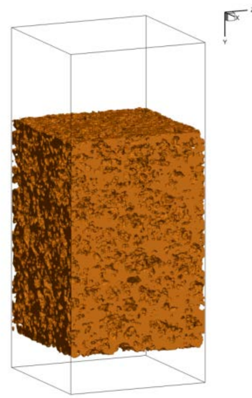
LB	lattice-Boltzmann
EFA	measurement
CFP	capillary flow porometry
PNM	pore network model
MPD	median pore diameter

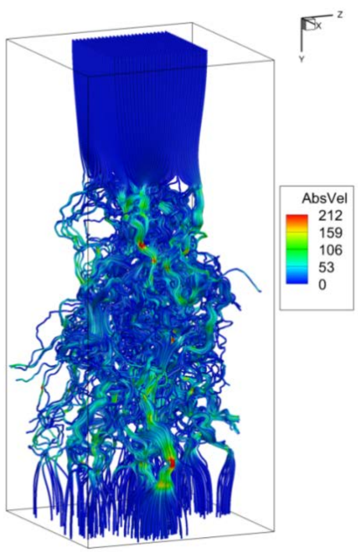
Analysis of filter with light coating

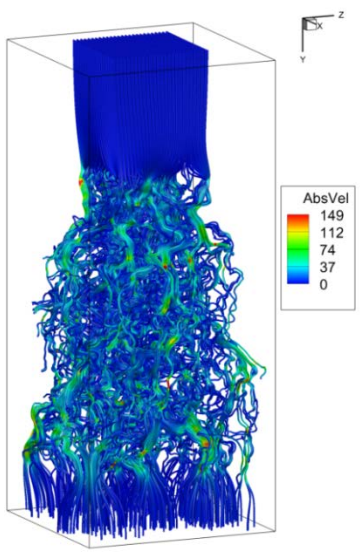












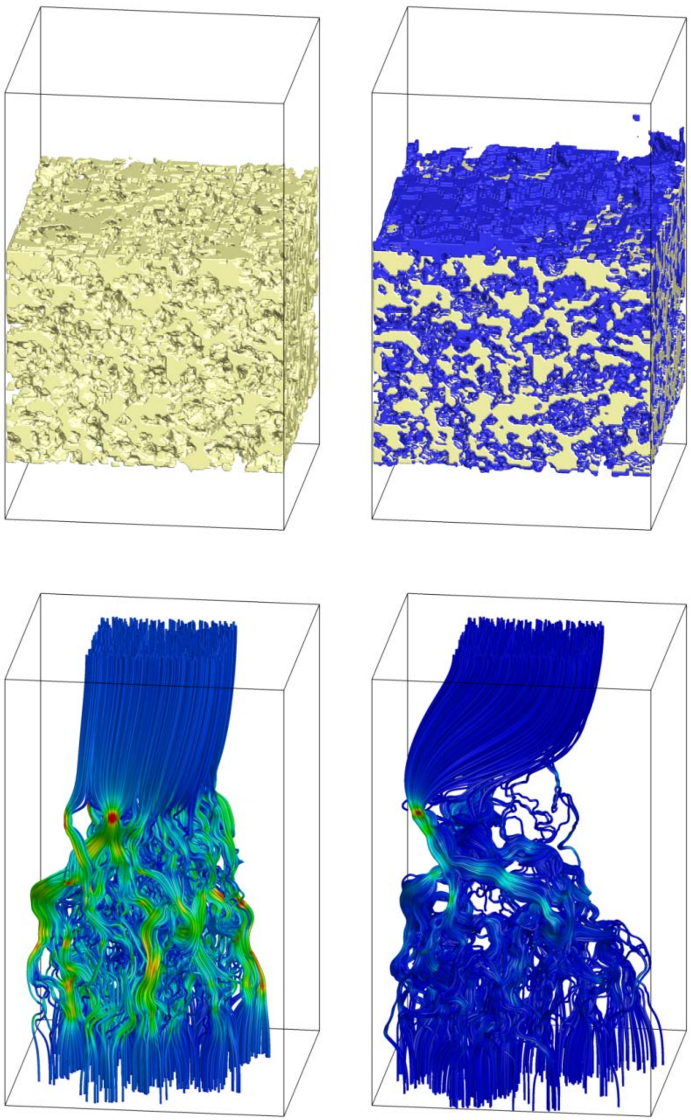
- Example: C1 filter wafer with oxidation catalyst
- No discrete catalyst phase discerned in CT data
- Geometric features, flow characteristics seem similar
- Catalyzed sample had larger pores - base substrate may not have been exactly the same

Very similar modeling approach likely applicable if the catalyzed sample is well characterized

Method	Parameter	Uncatalyzed	Catalyzed
Hg porosimetry	Porosity	43%	43%
	Median diam (μm)	12.0	13.4
	"W" metric	0.53	0.44
EFA	Permeability (m^2)	6.8±0.01E-13	5.0E-13*
Pore network model	Median body diam (μm)	16.0	17.4
	Median throat diam (μm)	11.4	12.7
	Permeability (m^2)	4.4E-13	1.2E-12
LB simulations	Permeability (m^2)	6.4E-13	1.1E-12
	Streamline tortuosity	1.62	1.60

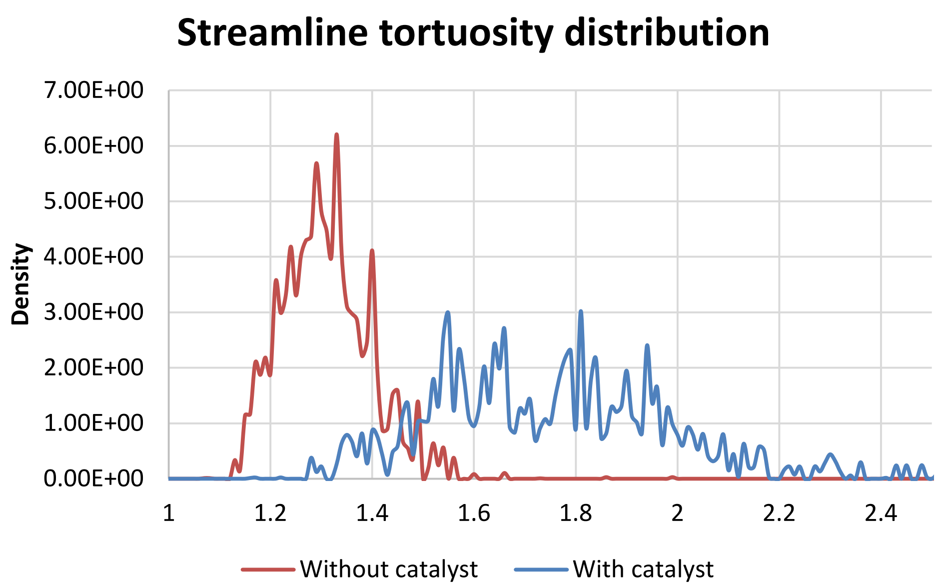
* preliminary measurement of a single sample

Technical accomplishments
LB simulations with heavy coating

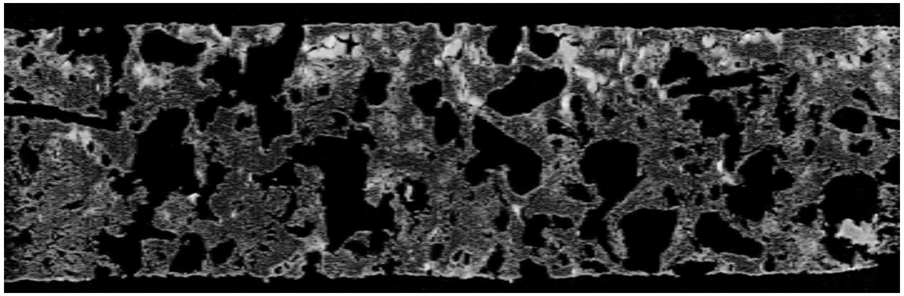


- Example: Flow field simulations with SCR-coated cordierite filter
- TWC-GPFs may also require relatively large washcoat volumes
- Heavy catalyst coatings alter the flow paths through the filter wall
- Depending on coating process, effective porosity and pore size may vary across the wall

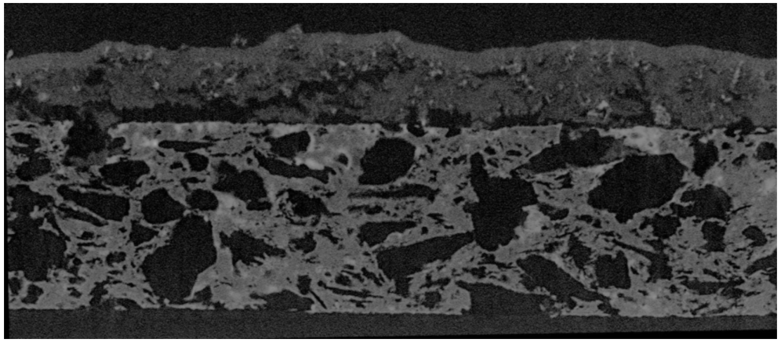
An ideal filter model would make appropriate adjustments to collector size, shape, connectivity



Extension to coated and ash-aged filters

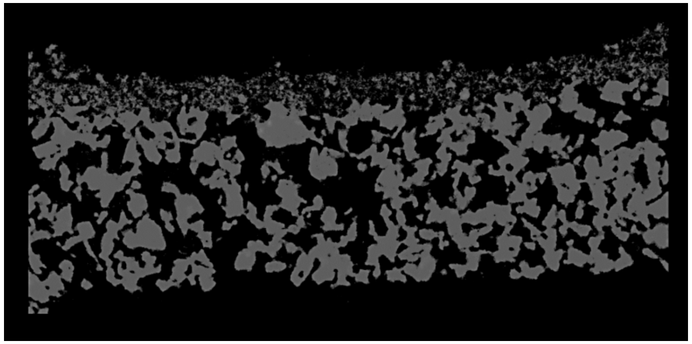


Coated cordierite - catalyst fills some pores at one wall surface



Coated cordierite with ash - some penetration into wall

MIT



SiC filter with ash layer

- High resolution (~1μm) and ultra-high resolution (~600-800nm) CT data has been obtained at MIT for an array of filter samples
  - Cordierite
  - Cordierite with oxidation catalyst
  - Cordierite with oxidation catalyst and 42 g/L ash
  - SiC with 20 g/L ash
  - Cordierite SCR-filter with ash
- Advanced segmentation methods will be used to generate 3D geometries
- Lattice-Boltzmann simulations will be performed and permeability predictions compared to experimental measurements
- Will also examine pore tortuosity/connectivity, etc.

EFA filtration experiments

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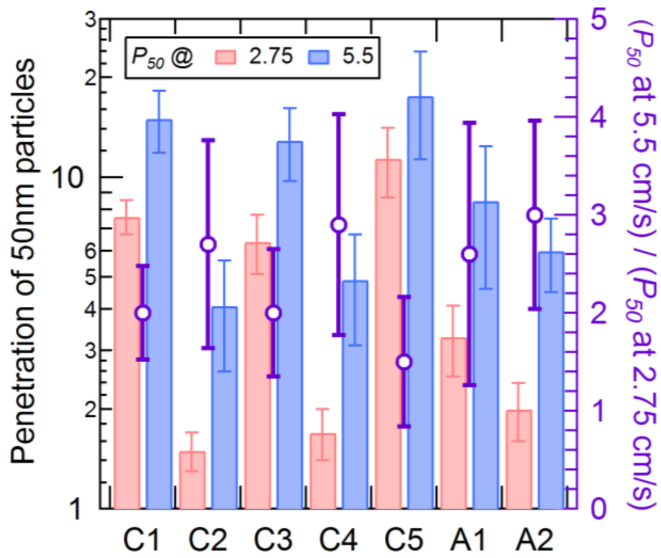
- Size-resolved filtration efficiency and pressure drop were measured as a function of loading using SIDI exhaust at two different wall velocities
- Example comparisons:
  - C2 and C4 similar except that C4 has a narrower pore size distribution
  - C3 and C4 similar except that C3 has much larger pores

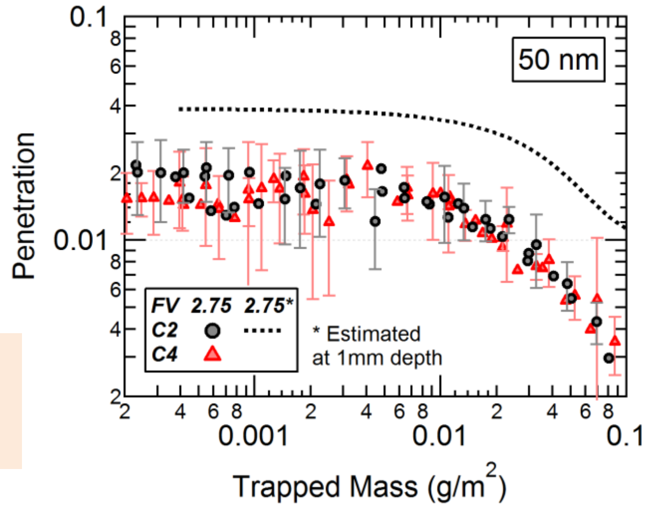
Large volume of fundamental filtration data collected. Currently being used to validate new models.

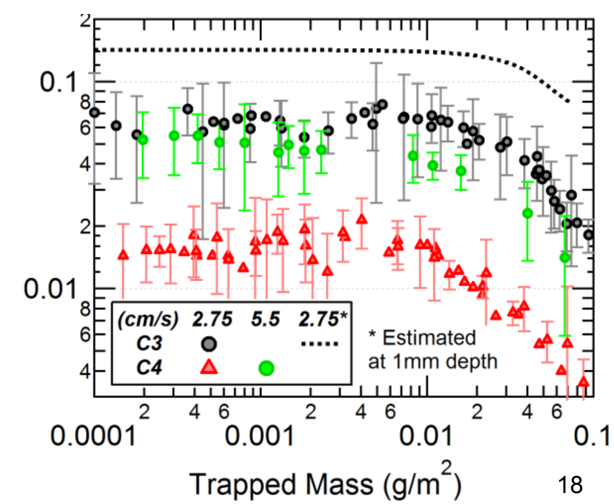
Viswanathan, S. *Experimental investigation of deep-bed filtration of spark-ignited, direct-injection engine exhaust using ceramic particulate filters.* **Doctoral dissertation.** University of Wisconsin, Madison, 2016.



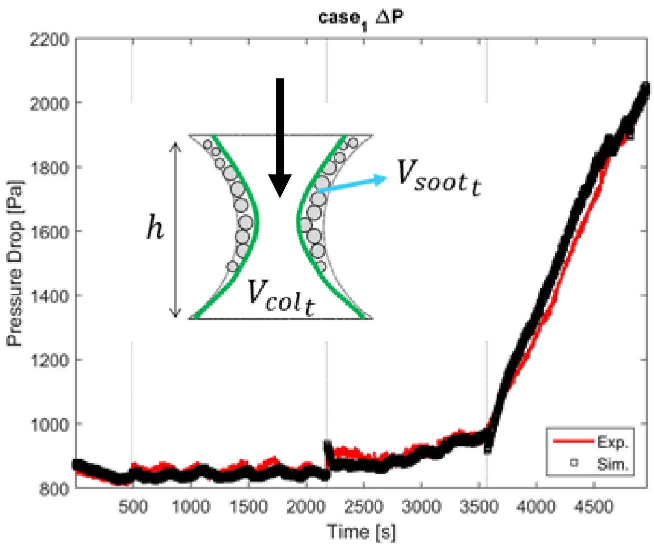
Example comparisons:

(* Dashed lines scaled for comparison of data from samples with different thickness)

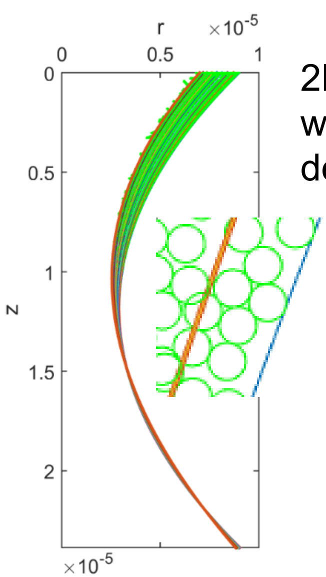




Constricted tube model development



- Work continues on a new wall-scale filtration model at U of Wisconsin, Madison - based on constricted tube collector
- Comparing predictions to large set of filtration data for various substrates, engine conditions
- Geometric evolution with pore filling based on 2D Lagrangian particle collection simulation
- 3D lattice-Boltzmann simulations are being used to look for more realistic functional forms



2D Lagrangian simulation without interaction between deposits and flow field

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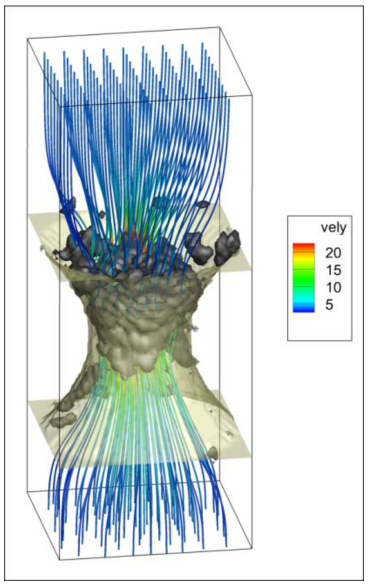
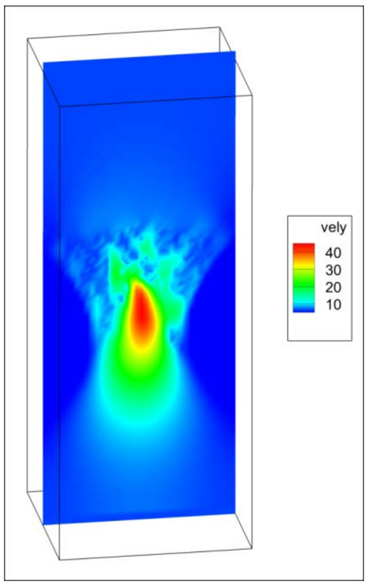
3D LB Lagrangian simulation with interaction between deposits and flow field





FY16 reviewer comments

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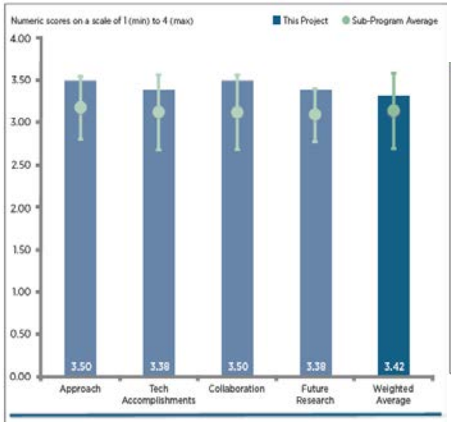

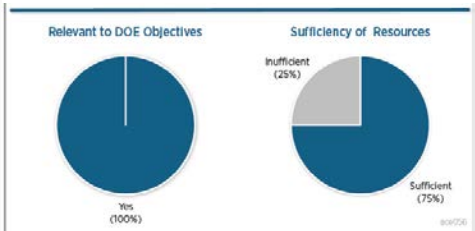

Figure 4-24 - Fuel-Neutral Studies of Particulate Matter Transport Emissions: Mark Stewart (Pacific Northwest National Laboratory) - Advanced Combustion Engines

- **"… this is a very important research project as OEMs are looking for data to help support development and implementation of GPFs for vehicles."**
- **"… there was an excellent use of advanced tools on a wide range of filters."**
- **"… found the overall research accomplishments to be fantastic and very relevant."**
- **"The Lattice Boltzmann simulations are quite interesting and explain much of the porosity impact on back pressure and filtration efficiency."**
- **"The reviewer remarked that there was good collaboration with GM and UW on this project."**
- **"…interaction with UW and GM has been very productive."**
- **"…this is an excellent list of needed future work on this project."**

- "… it is very important to understand the fuel properties of both splash and match-blended fuels, so it would be helpful to list that in the data for the project."

  All ethanol blends were splash-blended, but experiments were also carried out with various fuel surrogate blends which showed the impact of aromatic content on soot formation (see backup slide for example).

- "Their (experimental data) application into modeling, where appropriate, would be great to see more of."

  Our emphasis has continued to shift to model development in FY17.

- Effects of catalysts and ash should be considered.

  Catalyzed GPFs have been difficult to obtain for this kind of open research project, possibly due to impending introduction of commercial products. Work was started with a filter sample having an oxidation catalyst coating. MIT also joined the collaboration, with a view toward examining the effects of a range of catalyst coatings, as well as ash. Efforts will continue to obtain realistic TWC/GPF samples.

Collaborations

- Major Partners
  - General Motors Company (Industry): Provide funding (supporting full-time graduate student working on improved models), hardware, expertise, and operational guidance for work at the University of Wisconsin, Madison. Advise on project direction and priorities.
  - Engine Research Center at University of Wisconsin, Madison (Academic): Operate test engine and EFA system - including shakedown tests, independent experiments, and cooperative experiments. Assist in analysis and publication of data. Develop improved device-scale modeling techniques.
  - Massachusetts Institute of Technology (Academic): Perform X-Ray CT scans, including ultra-high (sub-micron) resolution. Provide access to datasets for catalyzed and ash-aged parts. Capabilities also include advanced artifact reduction and segmentation tools.

- Analysis subcontracts
  - Micromeritics
  - Particle Tech Labs
  - Micro Photonics

- Filter suppliers
  - Corning Incorporated
  - Ibidin
  - NGK
  - Sumitomo

Remaining challenges and barriers

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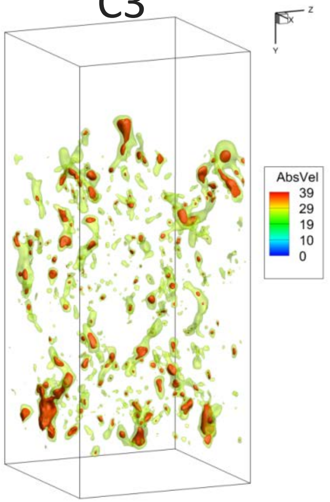
- Readily available characterization tools such as mercury porosimetry seem inadequate to completely describe the structural features of filters that determine performance.
- Rich datasets available from 3-D imaging show clear differences in the passages available for flow through various filter substrates and filters with various configurations of catalyst coatings and accumulated ash.

  Better ways must be found to translate these differences into quantifiable parameters that can be practically obtained for new or proposed filter products.
- More general models are needed, which will allow prediction of filter performance as a function of well-defined structural properties over a wide range of engine operating conditions, especially for the removal of the very small particles at low PM loadings expected for gasoline applications.
- Filtration data for continuous regeneration conditions at high temperature and low PM loading (representative of close-coupled GPF) is needed to validate relevant models.

C3



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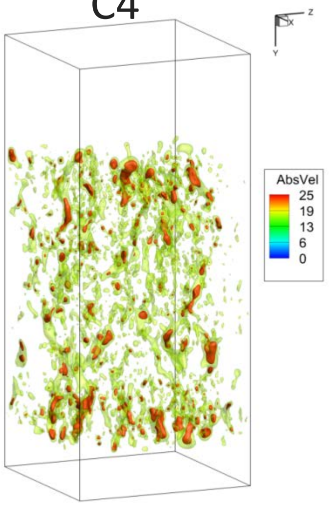

Future work

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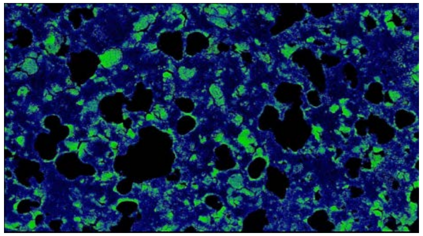
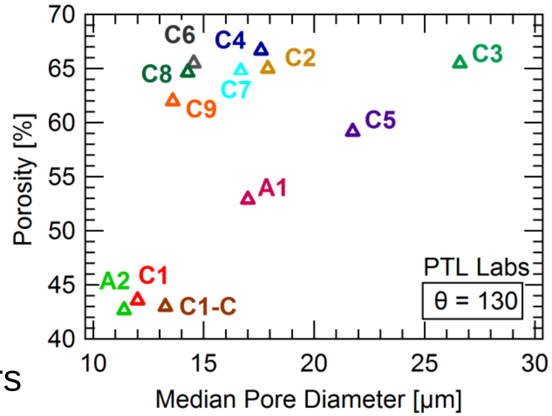


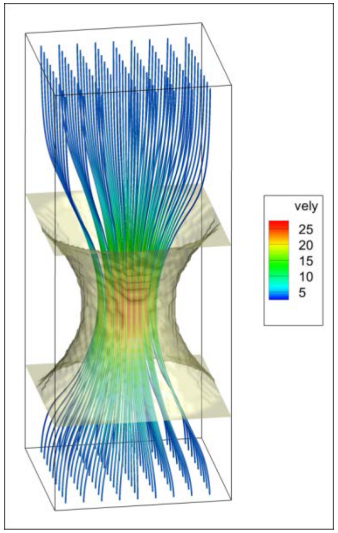
- Filer permeability prediction study in collaboration with MIT
  - Variety of catalyst coated and ash aged samples
  - Range of CT resolution, down to sub-micron
  - Advanced artifact reduction and segmentation techniques
  - Large domain, massively parallel lattice-Boltzmann simulations, possibly including sub-grid Darcy resistance model
- Additional low-temperature filtration studies at U of Wisconsin, Madison
  - Catalyzed filter samples
  - More uncatalyzed samples with thinner walls, high porosity, small pores
- Identify geometric descriptors of filter microstructure for more accurate predictions of pressure drop and filtration performance for bare substrates, ash-aged and coated filters
  - Path length through media
  - Pore shape / aspect ratio
  - Participating vs. non-participating pore volume
- Perform high temperature filter experiments representative of close-coupled placement
- Develop improved filtration models
  - Constricted tube unit collector
  - Enhanced spherical unit collector - based on new geometric parameters

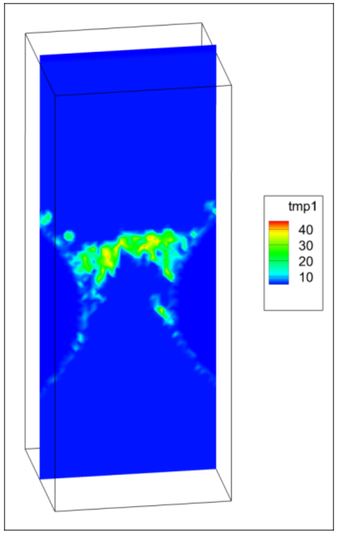


Any proposed future work is subject to change based on funding levels.

Summary

- Obtained high resolution X-Ray CT data for a variety of filters, including combinations of catalyst coatings and ash
- Applied new approaches for filter characterization
  - Experimental measurements
    - Capillary flow porometry
  - Examination of X-Ray CT data
    - Porosity distribution and uniformity
    - Pore network modeling
- Used detailed 3D lattice-Boltzmann simulations to probe effects of varying filter microstructure
  - Flow distribution
  - Flow path tortuosity
- Collected high quality fundamental filtration data
  - Seven different filter substrates covering a wide range of porosity, pore size, and permeability
  - Multiple flow rates
  - Multiple engine operating conditions / particulate populations
- Exploring alternatives to standard unit collector filtration model
  - Constricted tube unit collector model





Technical Back-Up Slides

Technical Accomplishments (FY 15)

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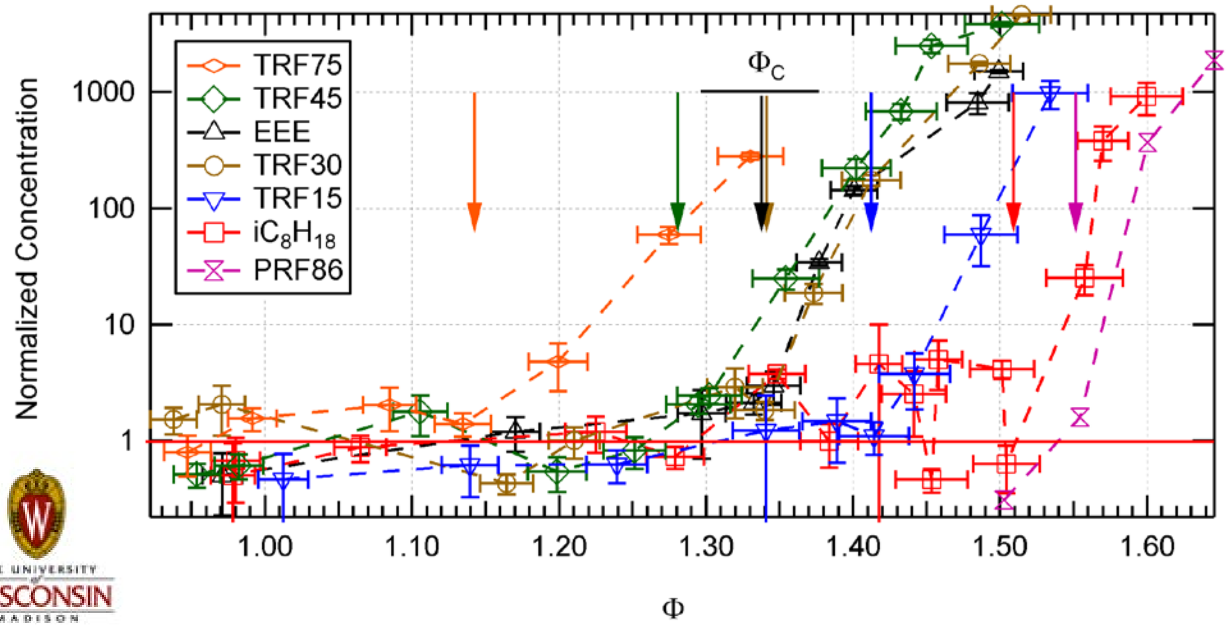
Fuel	Fuel C/H	Aromatics [Vol%]
EEE	0.583	28
TRF75	0.732	75
TRF45	0.597	45
TRF30	0.540	30
TRF15	0.489	15
PRF86	0.443	0
iC8H18	0.444	0

Particulate formation fundamentals - Critical equivalence ratios for soot



- Toluene reference fuel (TRF) blends used to probe effect of aromatic fraction during pre-mixed, pre-vaporized combustion
- Lower critical equivalence ratio (Φ_C) at higher aromatic fractions
- All observed Φ_C below the value of 2.0 observed with Diesel jets [1]
- Φ_C for TRF 30 similar to EEE

[1] L.M. Pickett, M. J., C.L. Genzale, D.L. Siebers, M.P.B. Musculus, C.A. Idicheria, SAE Technical Paper, 2011-01-0653 (2011).

Methods used to characterize filters

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Examples of major techniques used to characterize filter materials in this project and information provided by each

Mercury porosimetry	Pore size distribution → median pore size, W metric, log-normal standard deviation
Capilary flow porometry	Size distrubution of pore throats for through flow
	Clean permeability
EFA	Size-resolved filtration efficiency as a function of loading
	Pressure drop as a function of loading → clean permeability
X-ray CT data	Porosity distribution across wall thickness
	Porosity distribution over wall area (artifact check)
	Interior autocorrelation distribution (solid and void)
	Wall surface autocorrelation distribution (solid and void)
	Porosity representaive equivelent volume
	Solid chord length distributions (solid and void)
Pore network model	Pore body size distribution → median size
	Pore throat size distribution → median size, constriction ratio
	Pore throat length → pore aspect ratio, collector frequency through wall
	Por coordination number
	Clean permeability estimate
Lattice-Boltzman simulations	3D Flow field → Flow path tortuosity, connectivity
	Interstitial velocity distributions → Pore volume utilization for flow
	Size, freqency, connectivity of controlling restrictions for through-flow
	Clean permeability estimate
	Permeability representaive equivelent volume
	Estimate of removal efficiency for small particles (Eulerian simulation)
	Estimate of soot accumulation locations and pressure drop evolution (Lagrangian simulation)

Extensions of conceptual framework to coatings, ash

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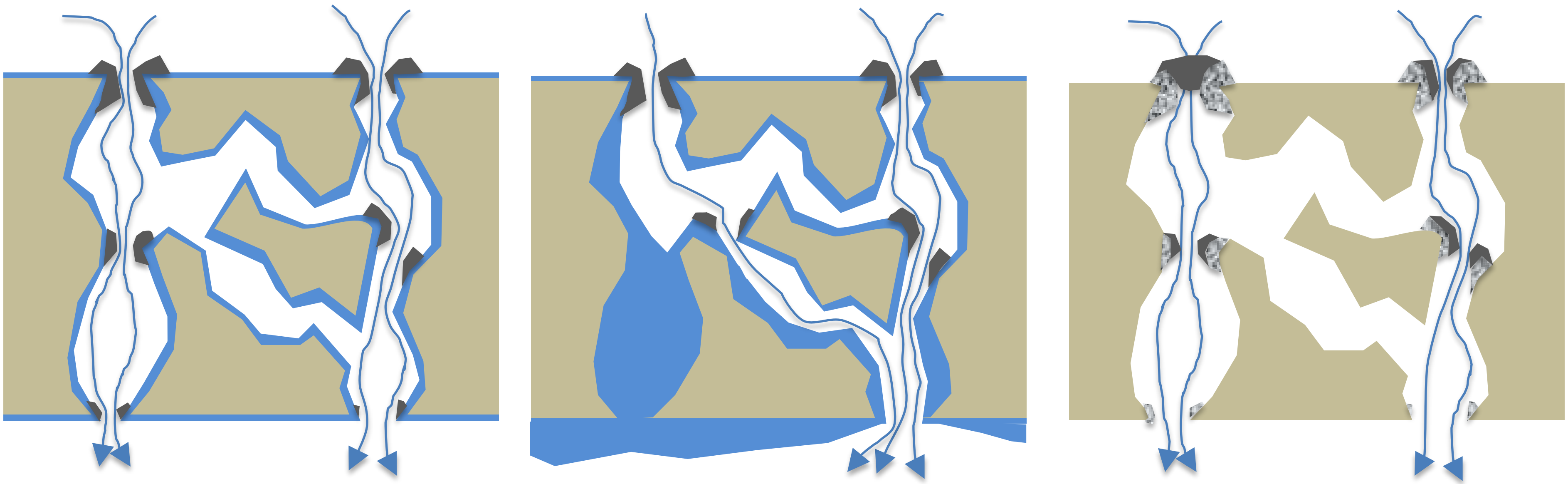
- Light, uniform coating
  - Examples: Some PGM-based oxidation catalysts
  - Some pores constricted slightly
  - Basic structure of pore network unchanged
- Heavy or non-uniform coating
  - Examples: SCR-filter, TWC-GPF?
  - Some pores filled or blocked
  - Pathways through filter wall altered
- Ash
  - Ash generated where soot is oxidized - more inside wall for GPF
  - Ash particles consolidate, migrate, form hydrates
  - Morphology varies, but ash typically porous, permeable



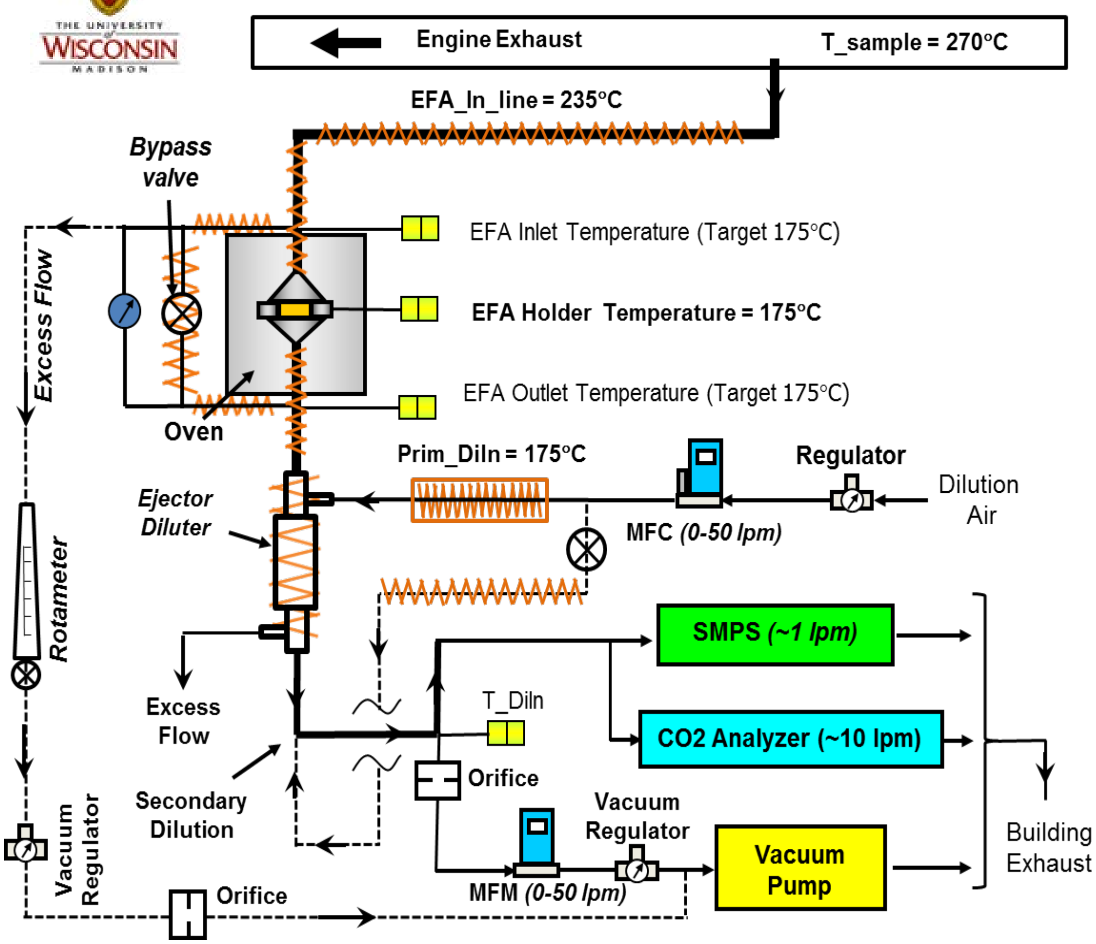
Ideally, the same general framework could be used to model various combinations, appropriately representing size and placement of controlling constrictions.

Exhaust Filtration Analysis (EFA) experiments

GM / UW-Madison Collaborative Research Laboratory

- Filtration experiments conducted with flat wafer samples and exhaust from single cylinder test engine
- Particulates measured with Scanning Mobility Particle Sizer (SMPS) and Engine Exhaust Particle Sizer (EEPS)

See SAE-2014-01-1558



Porosity representative volume analysis

- Re-ran porosity REV (representative equivalent volume) analysis after CT data had been adjusted to reduce gradient artifacts
- Looking at variation in porosity between sub-volumes of different sizes
- Whisker plot shows max and min values, 1st, 2nd, and 3rd quartiles